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PATENT SPECIFICATION

1,173,586

DRAWINGS ATTACHED.

1,173,586

Date of Application (No. 57831/66) and filing Complete Specification: 23 Dec., 1966.

Application made in Germany (No. H58104 V/37b) on 29 Dec., 1965.

Complete Specification Published: 10 Dec., 1969.

Index at acceptance:—B5 N(17Y, 17X, 171, 172, 176, 184, 22X, 221, 222, 239, 250, 254Y, 254X, 266Y, 265X, 266X, 280Y, 284Y, 285Y, 286Y, 298Y, 300X, 310X, 303X, 304X, 322Y, 326Y, 328, 329Y, 320, 324X, 326X, 329X, 333Y, 334Y, 335Y, 332X, 358, 371, 373, 397, 42X, 423, 424, 426, 428, 432, 436, 440, 445, 449, 45X, 507, 51X, 515, 540, 542, 546, 55Y, 591, 620, 63X, 64X, 669, 67X, 670, 671, 679, 68X, 682, 683, 69X, 71X, 711, 736, 74Y, 755, 768, 76X, 766, 768, 769, 79Y, 798, 801).

International Classification:—B 32 b 15/06.

COMPLETE SPECIFICATION.

Metal-Wood Composite Sheet.

We, HENKEL & CIE G.m.b.H., a German Company, of 67, Henkelstrasse, 4000 Duesseldorf-Holthausen, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to a composite sheet comprising metal and wood layers.

Composite sheets consisting of sheet metal and plywood layers are known. The bond between the said substances may be made by hot gluing with the aid of phenol resins. It is also possible to join plywood sheets and metal sheets together with the aid of contact adhesives, for example those based on natural or synthetic rubber. Moreover, it has become known to effect the bond between the materials by means of adhesives based on unsaturated polyesters or epoxide resins. The composite sheets obtainable by the said processes, however, have various disadvantages, such as the relatively brittle bond produced when phenol resins and unsaturated polyesters and frequently also epoxide resins are used. On the other hand, the strong adhesion often desired between wood and metal, for example aluminium, is not obtained with the contact adhesives. Further, difficulties occur during the preparation of such sheets in that contact adhesives and also adhesives based on unsaturated polyesters and epoxide resins must be worked within a specific time, due to their short pot-life.

The object of the invention is to provide a composite sheet comprising at least one metal layer and at least one layer of wood, which does not have the previously mentioned disadvantages.

According to the present invention there is provided a composite sheet having a metal layer, a layer of wood, and a layer of elastomer interposed between the metal layer and the wood layer and wherein the elastomer layer is joined to the metal layer and wood layer by an adhesive capable of bonding elastomer to metal.

The composition of the elastomer layer depends on the purpose for which the composite sheet is to be used. Suitable elastomers are natural rubber as well as a series of synthetic highly elastic products such as polychloroprene, polyisobutylene with small amounts therein of a polymerised conjugated diene, copolymers of butadiene and styrene or acrylonitrile, highly elastic copolymers of ethylene with vinyl acetate or propylene or also polyurethanes. The elastomer layer should preferably have a thickness of 0.1 to 10 mm., more preferably 0.2 to 2 mm.

The elastomer sheet may be used as such or a vulcanisable composition can be used and the composition vulcanised *in situ*.

Many different kinds of wood materials may be used as wood layers, for example joiner boards or plywood sheets. More-

over, it is possible to use chip boards. The wood layer should preferably have a thickness of 2 to 80 mm., more preferably 10 to 50 mm.

A larger variety of metal layers may be used in the composite sheet of the invention, such as iron, high-grade steel, copper, brass and zinc. However, aluminium sheets are preferred. The metal layers may be enamelled or, if desired, galvanised or chromium plated. They may have a coat of lacquer or designs may be embossed thereon. When iron sheets are used. they may be phosphatised on one or both sides. Aluminium sheets may be anodised or chromium plated on one or both sides. The metal layer should preferably have a thickness of 0.1 to 20 mm., more preferably 0.2 to 2 mm.

According to the invention, the elastomer layer is covered on both sides with a binding agent, which effects a firm bond between the elastomer layer and the metal layer and the wood sheet. For example, binding agents based on chlorinated rubber, polymers of chlorobutadiene and dichlorobutadiene, halogenated polymerisates of dichlorobutadiene, halogenated polymerisates of vinyl chloride or halogented copolymers of vinyl chloride and vinyl esters of lower carboxylic acids may be used for this purpose. It is also possible to use other commercial binding agents, for example rubber latices containing albumin or polyvalent isocyanates.

The layer of binding agent is preferably relatively thin. It preferably consists of 20 to 100 g/m^2, more preferably 30 to 80 g/m^2, of solids. For coating the layers, solutions of the binding agent in organic solvents or dispersions in water may be used. The solutions or dispersions may be applied in known way by brushes, sprays or rollers on the sides of the wood or metal layers which are facing the elastomer layer. After the solvent or water has evaporated, the layers can be combined to one composite.

The composite sheets according to the invention may contain a large variety of combinations of layers, comprising in the simplest case only one metal layer and one wood layer. Further, a wood sheet may be combined with two metal layers or a metal layer with two wood sheets. However, any desired number of layers can be bonded together, but in this case, an elastomer layer must always be present between metal and wood.

The construction of the composite sheet is started, for example, by coating the surface of the wood layer facing the elastomer layer with the adhesive. For this purpose plywood sheets previously coated with glue may be used or sheets consisting of plywood veneers which are partly coated with resin but not yet bonded may be used. The wood sheet facing the elastomer layer may be rough or have a thin layer of resin. In the latter case, less adhesive is required. Adhesive may also be saved by first placing a heavily resin-coated paper (the resin comprising about 150 to 250% of the weight of the paper), weighing about 15 to 40 g/m^2, on the rough side of the wood and then coating this with adhesive on the surface facing the elastomer layer. Suitable resins are the known, uncondensed, formaldehyde resins of phenol, cresol, melamine or urea.

The elastomer sheet and the metal layer are then applied to the wood sheet prepared as described above. It is also advantageous to coat the surface of the metal layer facing the elastomer with an adhesive, in addition to a coating of adhesive on the elastomer.

The bond between the separate layers is effected by compressing them over a period of about 5 to 30 minutes at a pressure of from 10 to 30 kg/cm^2 and at a temperature of from 120° to 180°C. At the temperature employed, a vulcanisation of the elastomer takes place. Since, instead of prepared plywood, various sheets of partly resin-coated wood veneers are preferably used, the bonding between individual wood layers takes place at the same time.

The invention is illustrated, by way of Example in the accompanying drawings.

In the drawing the metal-wood composite sheet is shown as comprising a metal sheet 1, a vulcanised rubber layer 2 and a veneered wood layer 3 consisting of individual wood veneer layers 6, 7 and 8. The rubber-metal interface 4 and the rubber-wood interface 5 have a thin layer of cured adhesive.

The composite sheets according to the invention have a very high bond strength between metal and wood. In most cases during a tensile test a break in the elastomer layer occurs, that is, the strength of the composite sheet is determined by the strength of the elastomer. Shear strengths of 50 kg/inch and over can be obtained.

The thin elastomer layer forms a flexible intermediate layer between the metal and wood layers and readily absorbs tensions of the composite sheet. Further, the elastomer layer acts as an equalising medium for any unevenness between the two materials. The composite sheet according to the invention is practically free from warping and has a high resistance against atmospheric influences and ageing.

The composite sheet according to the invention can be used advantageously as a cover plate for the interior and exterior construction of prefabricated houses, as lining for refrigeration plants, refrigerated transport vehicles, cooling tanks as well as in general for the construction of railway cars and carriages.

The invention will be further illustrated by reference to the following Examples.

Example 1

Five beechwood veneer sheets, of which two were coated on both sides with still hardenable phenolic resin and pre-dried, were placed on one another so that in each case the top and bottom sheets remained uncoated. One face of each of two smooth-rolled aluminium sheets of 0.4 and 0.2 mm thickness was washed with trichlorethylene and then coated with an adhesive capable of bonding rubber to metal so that an application of 40 g/m² of solids content was obtained. Also the side of the wood composite which was to face a rubber layer was coated with the adhesive (application 80 g/m² solids content).

The adhesive used was prepared in a known way (see German Patent 1,143,017). It consisted of 10 parts by weight of 4-dinitroso-diphenylamine, 30 parts by weight of brominated poly-2,3-dichlorobutadiene (about 27 per cent mol of bromine), 70 parts by weight of chlorinated natural rubber (125 centipoises) and 300 parts by weight of toluene.

An unvulcanised rubber sheet of about 0.5 mm thickness was inserted between the surfaces coated with the adhesive. The unvulcanised material consisted of about 100 g of polychloroprene, 2 g of phenyl-β-naphthylamine, 4 g of magnesium oxide, 1 g of a hydrocarbon resin, 5 g of petroleum softener, 5 g of zinc oxide, 70 g of lightly reinforcing carbon black, 0.5 g of di-o-tolyl-guanidine, 0.3 g of 2-mercaptonimidazoline, 0.5 g of tetramethylthiurammonosulphide, and 1.0 g of sulphur.

The layers were then compressed in a vulcanising press for 15 minutes at 150°C and a pressure of 20 kg/cm².

A firm composite body was obtained which had good mechanical strength.

The preparation of the composite body described above was repeated, but instead of the adhesive described, a commercial product with the trade name "Chemosil 220" (registered trade mark) was used. After a storage time of 24 hours, strips 1 inch in width were cut from the sheets thus obtained and the shear strength at an angle of 90° was measured across the length of 10 cm of a sample strip. The shear strength was from 41.1 kg/inch to 43.8 kg/inch. Up to 100% of the separation occurred in the elastomers. The test was conducted according to ASTM-D-429-55-T, Method B.

Example 2

Under the conditions given in Example 1, a composite sheet was prepared from a combination of five beechwood veneer layers and one aluminium sheet of 0.6 mm. thickness, lacquered on one side. The adhesive used consisted of 85 parts by weight of chlorinated natural rubber (a 20% solution in toluene had a viscosity of 125 centipoises at 20°C.), 15 parts by weight of a reaction product of a Novolac with epichlorhydrin (viscosity of 60,000 centipoises at 20°C., epoxide equivalent 179), 1.5 parts by weight of m-phenylenediamine and 30 parts by weight of finely divided carbon black.

Strips one inch in width were cut from the composite sheet and tested. The shear strength of the aluminium/wood composite amounted to 53.2 kg/inch, and separation occurred in the rubber layer over 95% of the whole adhesion surface. The following tests for stability were carried out according to DIN 68,705 and then the remaining shear strength was measured on strips one inch wide.

1. A 100: For testing the boiling resistance:
   Result: The composite was completely undamaged: shear strength 40.7 kg/inch, complete separation in the rubber layer.
2. AW 100: For testing the weather resistance of the composite:
   Result: The composite was entirely undamaged; shear strength 39.8 kg/inch, complete separation in the rubber layer.

Example 3

Of seven beachwood veneer sheets, four were coated with a phenolic resin and pre-dried. The veneer sheets were assembled into one composite of plywood, so that in addition to the glued layers both the top and bottom veneer sheets were pre-coated with resin. A commercial adhesive was applied to the top and bottom veneer sheets coated with phenolic resin (Chemosil 220). The coating comprised 50 g/m² of solids content. Two aluminium sheets 0.2 mm in thickness were washed with trichlorethylene and provided with a coating of about 40 g/m² of solids, of the same adhesive. Then an unvulcanised sheet of polychloroprene rubber of about 0.5 mm thickness was placed on each side of the plywood composite and on top of these the coated aluminium sheets were placed. The composite was vulcanised and bonded for 15 minutes at a temperature of 150°C. and under a pressure of 20 kg/cm².

A composite sheet was obtained from which strips one inch wide and 10 cm. long were cut. The shear strength amounted to 40 kg/inch at an angle of 90°, 100% separation occurring in the rubber.

The composite withstood the test for hot water resistance (IW 67 according to DIN 68,705) without damage. The remaining shear strength amounted to 36 kg/inch, and a separation of 80% was observed in the rubber.

Example 4

Seven beechwood veneer sheets, of which three were coated with adhesive, were combined to a composite, and a sodium kraft paper, saturated with a phenolic resin (35 g/m^2, 200% resinous) was placed thereon. This paper was coated on one side with a commercial adhesive (Chemosil 220, registered trade mark). After drying the adhesive, an unvulcanised rubber sheet 0.5 mm thick (as per Example 1) was placed thereon, and after that a 0.6 mm. thick aluminium sheet coated with the same adhesive (40 g/m^2). The composite thus obtained was bonded and vulcanised for 15 minutes at a temperature of 150°C. and under a pressure of 20 kg/cm^2.

Test strips one inch wide were cut from the composite sheet. The shear strength amounted to 48.2 kg/inch, and a 100% separation in the rubber layer was observed.

Example 5

Five beechwood veneer sheets, two of which were coated on both sides with still hardenable phenolic resin and pre-dried, were placed on one another so that in each case the top and bottom sheets remained uncoated. A copper sheet 0.5 mm thick was brushed and then coated on one side with an adhesive according to Example 2, so that an application of 20 g/m^2 of solids was obtained. One side of the wood composite was coated with the same adhesive (application of 60 g/m^2 of solids).

An unvulcanised rubber sheet according to Example 1 was placed between the coated surfaces and the vulcanisation was carried out for 15 minutes at a temperature of 150° C. under a pressure of 20 kg/cm^2.

A firm composite body was obtained which had excellent mechanical properties. In the tensile strength test, separation occurred in the rubber layer.

The above experiment was repeated, the copper sheet being replaced by a 0.3 mm. thick chromium-nickel-molybdenum-steel sheet. A firm composite was also obtained in which separation occurred in the rubber in the tensile test.

The est according to DIN 68,705 for boiling resistance and weather resistance was carried out on both composite sheets. Both sheets complied with the required conditions.

Example 6

Five beechwood veneer sheets were prepared according to Example 1 for the preparation of a composite sheet, and coated with an adhesive. The adhesive according to Example 1 was used. Smooth-rolled aluminium sheets of 0.4 and 0.2 mm thickness were cleaned and coated with the same adhesive.

An unvulcanised rubber layer of about 0.5 mm. thickness was inserted between the coated surfaces. The unvulcanised rubber material consisted of 100 parts of smoked sheet, 2 parts of stearic acid, 15 parts of zinc oxide, 2 parts of pine tar, 1 part of phenyl-β-naphthylamine, 25 parts of carbon black, 0.2 parts of zinc dimethyldithiocarbamate, 1.5 parts of mercaptobenzothiazyl-disulphide and 2.75 parts of sulphur.

The composite body was subsequently compressed in a vulcanising press for 10 minutes at a temperature of 150°C and under a pressure of 20 kg/cm^2.

A firm composite body was obtained which had good mechanical strength. In tensile tests, separation occurred in the rubber layer.

Example 7

Example 1 was repeated, but instead of the unvulcanised substance described therein, an unvulcanised rubber sheet of about 0.5 mm thickness and the following composition was used: 100 parts of styrene-butadiene copolymer indicated as SBR 1500 according to ASTM D 1419, 50 parts of carbon black, 5 parts of zinc oxide, 1 part of stearic acid, 8 parts of asphaltic resin, 1.25 parts of N-cyclohexyl-2-benzothiazyl-sulphonamide and 1.75 parts of sulphur.

The vulcanisation was carried out for 20 minutes at a temperature of 150°C. and under a pressure of 20 kg/cm^2. A firm composite body was obtained which satisfied the requirements according to DIN 68,705.

Example 8

Example 1 was repeated but instead of the unvulcanised material used therein, an unvulcanised rubber sheet of 0.5 mm. thickness was used which has the following composition: 100 parts of nitrile rubber (acrylonitrile content about 23%), 1 part of stearic acid, 10 parts of liquid terpene resin, 10 parts of dibutyl phthalate, 10 parts of zinc oxide, 75 parts of carbon black, 0.32 parts of tetramethylthiurammonosulphide and 1 part of sulphur.

The vulcanisation was carried out for 20 minutes at a temperature of 150°C. and under a pressure of 20 kg/cm^2. (A firm composite body was obtained. In the tensile test, separation occurred in the rubber.

Example 9

Example 2 was repeated, but instead of the rubber mixture used therein, an unvulcanised rubber sheet was used which had the following composition: 100 parts of butyl rubber (2 per cent mol of isoprene), 1 part of stearic acid, 5 parts of zinc oxide, 50 parts of carbon black, 1 part of mercaptobenzothiazyldisulphide, 1.5 parts of tellurium diethyldithiocarbamate and 1 part of sulphur.

The vulcanisation was carried out for 20 minutes at a temperature of 150°C. and under a pressure of 20 Kg/cm². A firm composite body was obtained. In the tensile test, separation occurred predominantly in the rubber layer.

WHAT WE CLAIM IS:—

1. A composite sheet comprising a metal layer, a layer of wood, and a layer of elastomer interposed between the metal layer and thte wood layer and wherein the elastomer layer is joined to the metal layer and wood layer by an adhesive capable of bonding elastomer to metal.

2. A composite sheet as claimed in claim 1 in which the elastomer layer is 0.1 to 10 mm. in thickness.

3. A composite sheet as claimed in claim 2 in which the elastomer layer is 0.2 to 2 mm. in thickness.

4. A composite sheet as claimed in any preceding claim in which the wood layer is 2 to 80 mm. in thickness.

5. A composite sheet as claimed in claim 4 in which the wood layer is 10 to 50 mm. in thickness.

6. A composite sheet as claimed in any preceding claim in which the metal layer is 0.1 to 20 mm. in thickness.

7. A composite sheet as claimed in claim 6 in which the metal layer is 0.2 to 2 mm. in thickness.

8. A composite sheet as claimed in any preceding claim in which the metal layer consists of aluminium.

9. A composite sheet as claimed in any preceding claim in which the adhesive comprises 20 to 100 g/m², of solids.

10. A composite sheet as claimed in claim 9 in which the adhesive comprises 30 to 80 g/m² of solids.

11. A composite sheet as claimed in any preceding claim which comprises more than one layer of metal and/or more than one layer of wood with a layer of elastomer between each pair of metal and wood layers.

12. A process for the preparation of a composite sheet as defined in any one of the preceding claims which comprises assembling a layer of wood, a metal layer and a layer of elastomer between the layer of metal and the layer of wood, the layer of elastomer being coated on each surface with an adhesive capable of bonding elastomer to metal and compressing the assembly for 5 to 30 minutes at a pressure of from 10 to 30 kg/cm² and at a temperature of from 120° to 180°C.

13. A process as claimed in claim 12 in which the layer of metal and/or the layer of wood is coated with said adhesive.

14. A process as claimed in claim 13 in which the wood layer consists of several wood veneer sheets partly coated with hardenable synthetic resins.

15. A process as claimed in claim 12 in which one side of the wood is covered with a resin-coated paper.

16. A process as claimed in any of claims 12 to 15 in which more than one layer of metal and/or more than one layer of wood are assembled with an elastomer layer between each pair of metal and wood layers.

17. A composite sheet as claimed in claim 1 substantially as hereinbefore described with reference to the foregoing Examples 1 to 4.

18. A composite sheet as claimed in claim 1 substantially as hereinbefore described with reference to the foregoing Examples 5 to 9.

19. A composite sheet as claimed in claim 1 substantially as hereinbefore described with reference to the accompanying drawing.

20. A process as claimed in claim 12, substantially as hereinbefore described with reference to the foregoing Examples 1 to 4.

21. A process as claimed in claim 12, substantially as hereinbefore described with reference to the foregoing Examples 5 to 9.

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Chartered Patent Agents.

Printed for Her Majesty's Stationery Office by Burgess & Son (Abingdon), Ltd.—1969.
Published at The Patent Office, 25 Southampton Buildings, London, W.C.2,
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1173586 COMPLETE SPECIFICATION

1 SHEET This drawing is a reproduction of the Original on a reduced scale